Relaxation theory of spin-3/2 Ising system near phase transition temperatures*

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Dynamics of a spin-3/2 Ising system Hamiltonian with bilinear and biquadratic nearest-neighbour exchange interactions is studied by a simple method in which the statistical equilibrium theory is combined with the Onsager's theory of irreversible thermodynamics. First, the equilibrium behaviour of the model in the molecular-field approximation is given briefly in order to obtain the phase transition temperatures, i.e. the first- and second-order and the tricritical points. Then, the Onsager theory is applied to the model and the kinetic or rate equations are obtained. By solving these equations three relaxation times are calculated and their behaviours are examined for temperatures near the phase transition points. Moreover, the z dynamic critical exponent is calculated and compared with the z values obtained for different systems experimentally and theoretically, and they are found to be in good agrement.

Keywords: spin-3/2 Ising system, Onsager theory

PACC: 0550, 7510H, 6460, 6460H, 6470, 0570L, 0570J

1. Introduction

The spin-3/2 Ising model Hamiltonian with bilinear (J) and biquadratic (K) nearest-neighbour exchange interactions, which is also known as the spin-3/2 isotropic Blume–Emery–Griffiths (BEG) model on the analogy of the spin-1 isotropic BEG model, has attracted much attention since it was originally introduced to explain the phase transition in $DyVO_4$ and its phase diagram was obtained in the molecular-field approximation (MFA).[1] Another spin-3/2 Ising model was later introduced to study tricritical properties in ternary fluid mixtures, which was also solved in MFA.[2] A spin-3/2 Ising model with the nearest-neighbour exchange interactions, both bilinear and biquadratic, and with a crystal-field interaction or a single-ion potential (D) is known as the spin-3/2 BEG model, which is an extension of the spin-1 BEG model.[3] Equilibrium properties of the spin-3/2 BEG model have been studied by well-known methods in equilibrium statistical physics such as the MFA and Monte Carlo (MC) simulation, a renormalization-group (RG) calculation, Honmura–Kaneyoshi differential operator technique and the effective field theory (EFT) based on the use of a probability distribution that currently accounts for the single-site kinematic relations and a cluster variational method in pair approximation.[4−8] An exact formulation of the spin-3/2 BEG system on a Bethe lattice was also studied by using the exact recursion relation.[9] Recently, the equilibrium behaviour of the antiferromagnetic spin-3/2 BEG model was also studied and its phase diagram was presented in Ref. [10]. Moreover, the ground state of the spin-3/2 BEG model was also studied in more detail.[11]

On the other hand, the spin-3/2 Blume–Capel (BC) model is a spin-3/2 Ising model Hamiltonian with only J and D interactions. The spin-3/2 BC model is also an extension of the spin-1 BC model,[12,13] which was first introduced by Blume and independently by Capel. The equilibrium behaviour of the spin-3/2 BC model has also been investigated by a variety of techniques in equilibrium statistical physics such as the MFA, the pair approximation, MC simulations, the transfer-matrix finite-size-scaling (TMFSS) calculations, a thermodynamically self-consistent theory based on an Ornstein–Zernike approximation, the Bethe–Peierls approximation (or the correlated effective field treatment), an effective field theory based on the use of a probability distribution, two-spin cluster approximation in the cluster expansion method.[14−16] The exact solutions of the ferromagnetic and the antiferromagnetic spin-3/2 BC models were presented on the Bethe lattice by means of the exact recursion relations.[17,18]

While much work has been done on the equilib-

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rium properties of the spin-3/2 Ising systems, there have been only a few investigations devoted to the nonequilibrium behaviour of the systems. An early attempt to study the dynamic aspect of the spin-3/2 BC model was made by Grandi and Figueiredo.[19] They employed the MC simulations and short-time dynamic scaling to determine the static and the dynamic critical exponents for the two-dimensional spin-3/2 BC model and discussed critical exponents, extensively. Keskin *et al.* studied dynamic phase transitions in the kinetic spin-3/2 BC,[20] spin-3/2 isotropic BEG[21] and spin-3/2 BEG[22] models in the presence of a time varying magnetic field by using the Glauber-type stochastic dynamics and presented the phase diagram in the (T, h) plane. They also obtained the dynamic phase diagrams of kinetic spin-3/2 Ising systems in (T, K) and (T, D) planes.[23,24] Moreover, the dynamic behaviour of the spin-3/2 isotropic BEG model was also studied by the path probability method (PPM) with point distribution.[25,26] Especially, the system approaching towards the stable and metastable equilibrium states was investigated. We also studied the time dependence of the spin-3/2 isotropic BEG model only near the second-order phase transition point or critical point[27] by the Onsager's theory of irreversible thermodynamics or Onsager reciprocity theorem (ORT).[28,29] Particularly, the relaxation of the order parameters and the behaviours of the relaxation times were investigated near the critical points. In our recent letter,[27] we did not study many aspects of the dynamics of the spin-3/2 isotropic BEG system due to the short and rapid communications. Therefore, the purpose of the present paper is to study the nonequilibrium critical phenomena of the system with the Onsager theory of irreversible thermodynamics or the ORT, extensively. Especially, we calculate the relaxation times and examine their behaviours near the first- and second-order phase transition points as well as the tricritical point. We also calculate the critical exponents for the relaxation times near the phase transition temperatures and the tricritical point numerically and compare them with the results obtained from the available models. The results of the present paper also lead us to study the critical behaviour of sound attenuation and dispersion in the spin-3/2 Ising system.

This type of calculation, namely the ORT, was first performed by Tanaka *et al.*[30] to study the relaxation phenomena in a spin-1/2 Ising system near the second-order phase transition point. Then Barry[31] and Barry and Harrington[32] raised the same problem for an AB-type ferromagnetic and antiferromagnetic spin-1/2 Ising model, respectively. Recently, Erdem and Keskin[33−35] used this type of calculation to study the dynamics of a spin-1 Ising system Hamiltonian with bilinear and biquadratic nearest-neighbour exchange interactions and they also studied the critical behaviours of the sound attenuation[36−38] and dispersion.[39] Erdem *et al.*[40] also investigated the relaxation phenomena in the spin-1 BEG model near the critical and the multicritical points by using the ORT. Moreover, very recently Gulpinar *et al.*[41] have used this method to study the dynamics of the iron group dihalides via the metamagnetic spin-1/2 Ising model.

We should also mention that the relaxation theory or phenomenon has attracted much attention in different systems, recently. For example, the relaxation processes of the two-dimensional XY model was studied by Lei and Zheng;[42] the relaxation analysis of a quasi-one-dimensional frustrated XY model was made by Nogawa and Nemoto;[43] the spin relaxation in $La_{1-x}Ca_xMnO_3$ was investigated by Atsarkin *et al.*;[44] the relaxation behaviour of spin glasses was investigated with in-field relaxation measurements by Etzkorn *et al.*[45] Moreover, the relaxation process in the photoinduced neutral-ionic paraelectric-ferroelectric phase transition in tetrathiafulvalene-p-chloranil;[46] the relaxations process in the antiferromagnetic Ising model on a triangular lattice;[47] the magnetic relaxation in antiferromagnetic $CoRh_2O_4$ nanoparticles;[48] the relaxation processes in PbSe quantum dots of different sizes[49] were also investigated.

The remaining part of this paper is organized as follows. In Section 2, the description of the model and its static properties are presented briefly. In Section 3, Onsager's theorem is briefly reviewed and the derivation of the kinetic or rate equations is given. In Section 4, the solution of the kinetic equations is presented near the equilibrium states. Finally, a summary and discussion of the results are given in the last section.

2. Model and equilibrium properties

The Hamiltonian of the spin-3/2 Ising system with bilinear and biquadratic interactions, also known as the isotropic spin-3/2 Blume–Emery–Griffiths

(BEG) model, is

$$H = -J\sum_{\langle ij\rangle} S_i S_j - K\sum_{\langle ij\rangle}\left(S_i^2 - \frac{5}{4}\right)\left(S_j^2 - \frac{5}{4}\right), \quad (1)$$

where the spins located at each site i on a discrete lattice can take the values $\pm 3/2$ and $\pm 1/2$, J and K are the nearest-neighbour bilinear and biquadratic exchange constants, respectively. It is a four-state three-order parameter system. The three order parameters are introduced as follows: (i) average magnetization ($M \equiv \langle S_i\rangle$), also called dipolar order parameter, (ii) quadrupolar parameter which is a function of the average of squared magnetization, i.e., $Q \equiv \langle S_i^2\rangle - 5/4$, (iii) octupolar order parameter which is a function of the average of cubed magnetization, i.e. $R \equiv 5\langle S_i^3\rangle/3 - 41\langle S_i\rangle/12$. These definitions of quadrupolar and octupolar order parameters ensure that $Q = 0.0$ and $R = 0.0$ at infinite temperature are different from the definitions $Q \equiv \langle S_i^2\rangle$ and $R \equiv \langle S_i^3\rangle$ used by many researchers.

In the molecular-field approximation in the Bragg–Williams formalism, the Helmholtz free energy of the spin-3/2 Ising system with bilinear and biquadratic interactions is given as

$$F = -NJM^2 - NKQ^2 - kT\ln W(M,Q,R), \quad (2)$$

where N, k and T are the number of Ising spins, the Boltzmann factor and the absolute temperature, respectively, and $W(M,Q,R)$ is the number of configurations for given values of dipolar, quadrupolar and octupolar order parameters. The last term in Eq. (2) is given by

$$\begin{aligned}&\ln W(M,Q,R)\\ &= -N\Bigg[\left(\frac{1}{4}+\frac{3M}{10}+\frac{Q}{4}+\frac{R}{10}\right)\ln\left(\frac{1}{4}+\frac{3M}{10}+\frac{Q}{4}+\frac{R}{10}\right)+\left(\frac{1}{4}+\frac{M}{10}-\frac{Q}{4}-\frac{3R}{10}\right)\ln\left(\frac{1}{4}+\frac{M}{10}-\frac{Q}{4}-\frac{3R}{10}\right)\\ &\quad+\left(\frac{1}{4}-\frac{M}{10}-\frac{Q}{4}+\frac{3R}{10}\right)\ln\left(\frac{1}{4}-\frac{M}{10}-\frac{Q}{4}+\frac{3R}{10}\right)+\left(\frac{1}{4}-\frac{3M}{10}+\frac{Q}{4}-\frac{R}{10}\right)\ln\left(\frac{1}{4}-\frac{3M}{10}+\frac{Q}{4}-\frac{R}{10}\right)\Bigg]. \quad (3)\end{aligned}$$

We minimize the Helmholtz free energy with respect to order parameters, and we obtain the following set of self-consistent equations for the equilibrium values of the order parameters:

$$M_0 = \frac{3\,\mathrm{e}^{2\beta KQ_0}\sinh(3\beta JM_0) + \mathrm{e}^{-2\beta KQ_0}\sinh(\beta JM_0)}{2\,\mathrm{e}^{2\beta KQ_0}\cosh(3\beta JM_0) + 2\,\mathrm{e}^{-2\beta KQ_0}\cosh(\beta JM_0)}, \quad (4)$$

$$Q_0 = \frac{2\,\mathrm{e}^{2\beta KQ_0}\cosh(3\beta JM_0) - 2\,\mathrm{e}^{-2\beta KQ_0}\cosh(\beta JM_0)}{2\,\mathrm{e}^{2\beta KQ_0}\cosh(3\beta JM_0) + 2\,\mathrm{e}^{-2\beta KQ_0}\cosh(\beta JM_0)}, \quad (5)$$

$$R_0 = \frac{\mathrm{e}^{2\beta KQ_0}\sinh(3\beta JM_0) - 3\,\mathrm{e}^{-2\beta KQ_0}\sinh(\beta JM_0)}{2\,\mathrm{e}^{2\beta KQ_0}\cosh(3\beta JM_0) + 2\,\mathrm{e}^{-2\beta KQ_0}\cosh(\beta JM_0)}, \quad (6)$$

where $\beta = 1/kT$. It should be mentioned that equations (4), (5) and (6) can be derived directly (with making allowance for the different notations) from the mean-field approximation via the variational principle for the free energy.[1,50,51] Since the solution of these equations was discussed extensively in Refs. [1], [50] and [51], we shall give only a brief summary here. For $J/K < 0.46$ a second-order phase transition occurs to a state with M and Q, and for $0.46 < J/K < 0.61$ a first-order phase transition occurs to a state with M and a second-order phase transition for Q. For $J/K < 0.61$ there is a quadrupolar region in the phase diagram. For $0.61 < J/K < 164/85$ a first-order phase transition and finally $J/K > 164/85$ a second-order phase transition appears again for M and Q. The system has two tricritical points at $J/K = 0.46$ and $J/K = 164/85$ respectively. This information is very important for studying the nonequilibrium critical phenomena of the system.

3. Theory

3.1. Summary of Onsager's reciprocity theorem

In order to state Onsager's theorem, we consider a system whose states can be described by a set of n independent variables A_i $(i = 1, 2, \ldots, n)$. An arbitrary state can be specified by the deviations α_i from

the equilibrium value (A_i^0), i.e.

$$\alpha_i = A_i - A_i^0. \tag{7}$$

The state variables α_i are zero by this definition. In the neighbourhood of an equilibrium state, the Helmholtz free energy is expanded into a Taylor series from deviations in the thermodynamic quantities and only up to the second order quantities are retained. In this approximation free energy is given by

$$F = F_0 + T\Delta S, \tag{8}$$

where F_0 is the equilibrium free energy, and ΔS is the entropy production that is given by

$$\Delta S = -\frac{1}{2}\sum_{i,j} g_{ij}\alpha_i\alpha_j, \qquad (i,j = 1,2,...). \tag{9}$$

Since entropy tends to increase to a maximum, entropy has a maximum value at the equilibrium. Therefore, g_{ij} are positive-definite matrix elements that are called entropy production coefficients and given by

$$g_{ij} = -\left(\frac{\partial^2 \Delta S}{\partial\alpha_i\partial\alpha_j}\right)_0 = \frac{1}{T}\left(\frac{\partial^2 F}{\partial\alpha_i\partial\alpha_j}\right)_0. \tag{10}$$

Let forces X_i and fluxes J_i be defined as

$$X_i = \frac{\partial\Delta S}{\partial\alpha_i} = -\sum_j g_{ij}\alpha_j, \tag{11}$$

$$J_i = \frac{\mathrm{d}\alpha_i}{\mathrm{d}t}. \tag{12}$$

Evidently, all J_i and X_i vanish at equilibrium. Then, the forces and the fluxes are related by the linear phenomenological equations

$$J_i = \sum_j L_{ij}X_j, \tag{13}$$

stating that any flow is caused by contribution of all forces. The L_{ij} coefficients are called phenomenological or kinetic coefficients. Onsager's reciprocal relation[28,29] states that

$$L_{ij} = \pm L_{ji}, \tag{14}$$

where the plus and the minus signs correspond to quantities α_i and α_j having the same parities and different parities under time reversal, respectively.

3.2. Derivation of kinetic equations

In order to derive the kinetic equations, let $A_1 = M$, $A_2 = Q$ and $A_3 = R$ be the dipolar, quadrupolar and octupolar order parameters in the spin-3/2 Ising system, respectively. Then the deviations of these parameters from their equilibrium values are given by

$$\begin{aligned}&\alpha_M = M - M_0, \ \ \alpha_Q = Q - Q_0,\\ &\alpha_R = R - R_0,\end{aligned} \tag{15}$$

where M_0, Q_0 and R_0 are the equilibrium values. Using Eq. (9), the entropy production for a nonequilibrium state in the system is written as

$$\begin{aligned}\Delta S = -\frac{1}{2}[&g_{MM}(M-M_0)^2 + 2g_{MQ}(M-M_0)(Q-Q_0) + 2g_{MR}(M-M_0)(R-R_0)\\ &+ g_{QQ}(Q-Q_0)^2 + 2g_{QR}(Q-Q_0)(R-R_0) + g_{RR}(R-R_0)^2]\end{aligned} \tag{16}$$

with coefficients defined by

$$g_{MM} = \frac{1}{T}\left(\frac{\partial^2 F}{\partial M^2}\right)_0 = -Nk\left[\frac{2J}{kT} - \frac{18(1+Q_0)}{d_1} - \frac{2(1-Q_0)}{d_2}\right], \tag{17a}$$

$$g_{MQ} = \frac{1}{T}\left(\frac{\partial^2 F}{\partial M\partial Q}\right)_0 = Nk\left[-\frac{6(3M_0+R_0)}{d_1} + \frac{2(M_0-3R_0)}{d_2}\right], \tag{17b}$$

$$g_{MR} = \frac{1}{T}\left(\frac{\partial^2 F}{\partial M\partial R}\right)_0 = Nk\left[\frac{6(1+Q_0)}{d_1} - \frac{6(1-Q_0)}{d_2}\right], \tag{17c}$$

$$g_{QQ} = \frac{1}{T}\left(\frac{\partial^2 F}{\partial Q^2}\right)_0 = -Nk\left[\frac{2K}{kT} - \frac{25(1+Q_0)}{2d_1} - \frac{25(1-Q_0)}{2d_2}\right], \tag{17d}$$

$$g_{QR} = \frac{1}{T}\left(\frac{\partial^2 F}{\partial Q\partial R}\right)_0 = Nk\left[-\frac{2(3M_0+R_0)}{d_1} - \frac{6(M_0-3R_0)}{d_2}\right], \tag{17e}$$

$$g_{RR} = \frac{1}{T}\left(\frac{\partial^2 F}{\partial R^2}\right)_0 = Nk\left[\frac{2(1+Q_0)}{d_1} + \frac{18(1-Q_0)}{d_2}\right], \tag{17f}$$

where $d_1 = (5+6M_0+5Q_0+2R_0)(5-6M_0+5Q_0-2R_0)$, and $d_2 = (5+2M_0-5Q_0-6R_0)(5-2M_0-5Q_0+6R_0)$. Using Eqs. (11), (15) and (17), we can obtain the forces as

$$\begin{pmatrix} X_M \\ X_Q \\ X_R \end{pmatrix} = \begin{pmatrix} -g_{MM} & -g_{MQ} & -g_{MR} \\ -g_{MQ} & -g_{QQ} & -g_{QR} \\ -g_{MR} & -g_{QR} & -g_{RR} \end{pmatrix} \begin{pmatrix} M-M_0 \\ Q-Q_0 \\ R-R_0 \end{pmatrix}. \tag{18}$$

According to the Onsager theory of irreversible thermodynamics, the linear relations between the currents and the forces given by Eq. (13) may be written as a matrix of phenomenological rate coefficients where the off-diagonal elements are the negatives of their corresponding elements symmetric with respect to the main diagonal (the matrix is not symmetric since M and R are odd variables while Q is an even variable under time inversion):

$$\begin{pmatrix} J_M \\ J_Q \\ J_R \end{pmatrix} = \begin{pmatrix} \gamma_M & -\gamma & \epsilon \\ \gamma & \gamma_Q & -\eta \\ \epsilon & \eta & \gamma_R \end{pmatrix} \begin{pmatrix} X_M \\ X_Q \\ X_R \end{pmatrix}. \tag{19}$$

Substituting Eq. (18) into Eq. (19) and using Eqs. (12) and (15), we obtain the kinetic equations as

$$\begin{pmatrix} \dot{M} \\ \dot{Q} \\ \dot{R} \end{pmatrix} = \begin{pmatrix} -\gamma_M g_{MM} + \gamma g_{MQ} - \epsilon g_{MR} & -\gamma_M g_{MQ} + \gamma g_{QQ} - \epsilon g_{QR} & -\gamma_M g_{MR} + \gamma g_{QR} - \epsilon g_{RR} \\ -\gamma g_{MM} - \gamma_Q g_{MQ} + \eta g_{MR} & -\gamma g_{MQ} - \gamma_Q g_{QQ} + \eta g_{QR} & -\gamma g_{MR} - \gamma_Q g_{QR} + \eta g_{RR} \\ -\epsilon g_{MM} - \eta g_{MQ} - \gamma_R g_{MR} & -\epsilon g_{MQ} - \eta g_{QQ} - \gamma_R g_{QR} & -\epsilon g_{MR} - \eta g_{QR} - \gamma_R g_{RR} \end{pmatrix} \times \begin{pmatrix} M-M_0 \\ Q-Q_0 \\ R-R_0 \end{pmatrix}. \tag{20}$$

The solution and the discussion of Eq. (20) are given in the next section.

4. Solution of kinetic equation near equilibrium states

In order to study the dynamics of the model in the neighbourhood of equilibrium states, a solution in the form of $\mathrm{e}^{-t/\tau}$ is assumed for the linearized kinetic equations given by Eq. (20) and the approaches of $M(t)$, $Q(t)$ and $R(t)$ to their equilibrium values are described by three characteristic times, which are also called the relaxation times. Therefore, to find these times the solution of the following secular equation is required:

$$\begin{vmatrix} \tau^{-1} - \gamma_M g_{MM} + \gamma g_{MQ} - \epsilon g_{MR} & -\gamma_M g_{MQ} + \gamma g_{QQ} - \epsilon g_{QR} & -\gamma_M g_{MR} + \gamma g_{QR} - \epsilon g_{RR} \\ -\gamma g_{MM} - \gamma_Q g_{MQ} + \eta g_{MR} & \tau^{-1} - \gamma g_{MQ} - \gamma_Q g_{QQ} + \eta g_{QR} & -\gamma g_{MR} - \gamma_Q g_{QR} + \eta g_{RR} \\ -\epsilon g_{MM} - \eta g_{MQ} - \gamma_R g_{MR} & -\epsilon g_{MQ} - \eta g_{QQ} - \gamma_R g_{QR} & \tau^{-1} - \epsilon g_{MR} - \eta g_{QR} - \gamma_R g_{RR} \end{vmatrix} = 0. \tag{21}$$

Solution of this determinant yields the following three inverse relaxation times:

$$\tau_1^{-1} = -\frac{b}{3a} + \frac{2}{3a}\sqrt{b^2-3ac}\cos\left(\frac{\theta+2\pi}{3}\right), \tag{22a}$$

$$\tau_2^{-1} = -\frac{b}{3a} + \frac{2}{3a}\sqrt{b^2-3ac}\cos\left(\frac{\theta}{3}\right), \tag{22b}$$

$$\tau_3^{-1} = -\frac{b}{3a} + \frac{2}{3a}\sqrt{b^2-3ac}\cos\left(\frac{\theta+4\pi}{3}\right), \tag{22c}$$

where

$$\begin{aligned} a &= 1, \\ b &= -\gamma_M g_{MM} - \gamma_Q g_{QQ} - \gamma_R g_{RR} - 2\epsilon g_{MR}, \\ c &= \gamma_M\gamma_Q(g_{MM}g_{QQ} - g_{MQ}^2) \\ &\quad + \gamma_M\gamma_R(g_{MM}g_{RR} - g_{MR}^2) \\ &\quad + \gamma_Q\gamma_R(g_{QQ}g_{RR} - g_{QR}^2) \\ &\quad + g_{MM}(\gamma^2 g_{QQ} - \epsilon^2 g_{RR}) \\ &\quad + g_{QQ}(\eta^2 g_{RR} - 2\gamma\eta g_{MR}) \end{aligned}$$

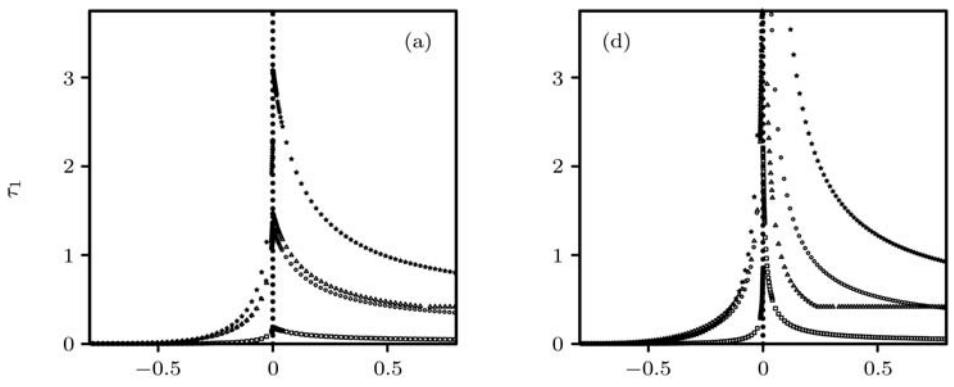
$$
\begin{aligned}
&+ 2\gamma_Q\epsilon(g_{MR}g_{QQ} - g_{MQ}g_{QR}) - \gamma^2 g_{MQ}^2 \\
&- \eta^2 g_{QR}^2 + \epsilon^2 g_{MR}^2 + 2\gamma\eta g_{MQ}g_{QR}, \\
d = {} & (\gamma_M\eta^2 - \gamma_Q\epsilon^2 + \gamma_R\gamma^2 + \gamma_M\gamma_Q\gamma_R)(g_{MM}g_{QR}^2 \\
&+ g_{QQ}g_{MR}^2 + g_{RR}g_{MQ}^2 - g_{MM}g_{QQ}g_{RR} \\
&- 2g_{MR}g_{MQ}g_{QR}) + 2\epsilon\gamma\eta g_{MM}(g_{QR}^2 - g_{QQ}g_{RR}) \\
&+ 2\epsilon\gamma\eta(g_{MR}^2 g_{QQ} + g_{MQ}^2 g_{RR} - 2g_{MQ}g_{MR}g_{QR})
\end{aligned}
$$

and

$$\theta = \arccos\left(\frac{-2b^3 + 9abc - 27a^2 d}{2\sqrt{(b^2 - 3ac)^3}}\right).$$

In this paper we made the assumption that the off-diagonal rate coefficients are negligible compared with diagonal ones for temperature close to the critical temperature. A similar assumption was also made in Refs. [30], [31] and [32] for spin-1/2, Refs. [33]–[40] for spin-1 Ising, and Ref. [41] for the metamagnetic spin-1/2 Ising models. Strictly speaking, such an assumption may only be completely verified by a theory external to irreversible thermodynamics.[31]

These three relaxation times manage to approach the equilibrium values of the order parameters, i.e. dipolar, quadrupolar and octupolar order parameters M, Q and R approach their equilibrium values M_0, Q_0 and R_0 with these three characteristic times τ_1, τ_2 and τ_3 given by Eqs. (22). It is expected from the conventional theory that there is at least one mode whose relaxation rate goes to zero at the critical point. This slow relaxation of the order parameter can lead to observable anomalies in dynamic properties.[52] One of the relaxation times (τ_1) increases rapidly with temperature and tends to infinity near the second-order phase transition temperature or critical temperature on both sides. Therefore, the motions of the order parameters slow down enormously near the critical point, strong attenuation and dispersion of sound are expected near that point as often observed experimentally.[53] We have observed that one of the relaxation times goes to infinity which is responsible for slow relaxation of the orders parameters and already presented the behaviour of the relaxation times each as a function of the temperature near the second-order phase transition or critical point, seen in Figs. 1 and 2 of Ref. [27]. In Figs. 1, 2 and 3, circles correspond to the case where $\gamma_M = 7.0, \gamma_Q = 9.0, \gamma_R = 4.0, \gamma = 0.04, \epsilon = 0.005$, and $\eta = 0.006$; triangles represent the case where $\gamma_M = 15.0, \gamma_Q = 5.0, \gamma_R = 3.0, \gamma = 0.001, \epsilon = 0.001$, and $\eta = 0.005$; stars denote the case where $\gamma_M = 3.0, \gamma_Q = 10.0, \gamma_R = 20.0, \gamma = 0.08, \epsilon = 0.010$, and $\eta = 0.0030$ and squares denote the case where $\gamma_M = 50.0, \gamma_Q = 75.0, \gamma_R = 80.0, \gamma = 0.01, \epsilon = 0.001$, and $\eta = 0.002$. Since the behaviours of the relaxation times at the second-order transition or critical temperatures are given and discussed in Ref. [27], we will no longer present and discuss these figures here. However we should explain the behaviour of relaxation times briefly. One of the relaxation times (τ_1) increases rapidly with temperature increasing and tends to infinity at the second-order phase transition point. Therefore, the motions of the order parameters slow down enormously near the critical point. The other relaxation time (τ_2) increases slightly with temperature increasing and makes a cusp at the second-order phase transition temperatures. The third relaxation time (τ_3) behaves very differently: it terminates at the second-order phase transition temperature while approaching it, then showing a "flatness" property and then decreases with temperature increasing. Moreover, we have also found that the behaviours of relaxation times at the tricritical point are the same as those in the second-order phase transition case; hence we do not present their behaviours again.



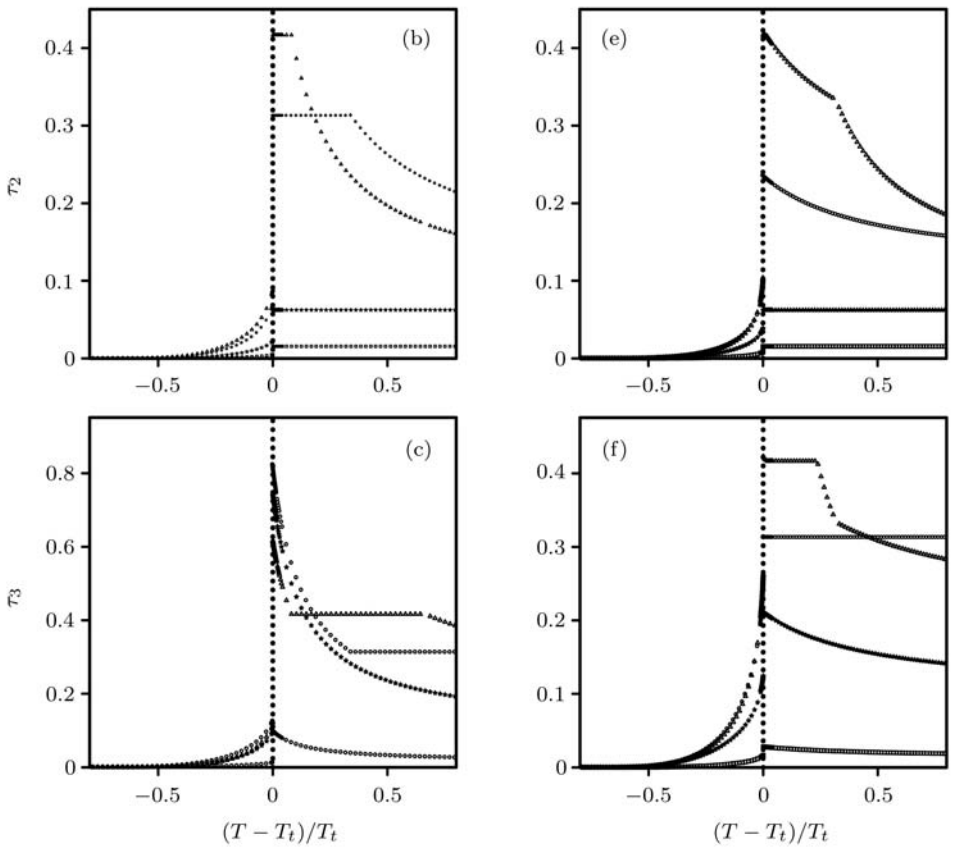


Fig. 1. Behaviours of relaxation times each as a function of reduced temperature. for $J/K = 0.8$ (a), (b) and (c), and $J/K = 1.5$ (d), (e) and (f), where circles correspond to $\gamma_M = 7.0, \gamma_Q = 9.0, \gamma_R = 4.0, \gamma = 0.04, \epsilon = 0.005$, and $\eta = 0.006$; triangles represent $\gamma_M = 15.0, \gamma_Q = 5.0, \gamma_R = 3.0, \gamma = 0.001, \epsilon = 0.001$, and $\eta = 0.005$; stars illustrate $\gamma_M = 3.0, \gamma_Q = 10.0, \gamma_R = 20.0, \gamma = 0.08, \epsilon = 0.010$, and $\eta = 0.0030$; squares indicate $\gamma_M = 50.0, \gamma_Q = 75.0, \gamma_R = 80.0, \gamma = 0.01, \epsilon = 0.001$, and $\eta = 0.002$.

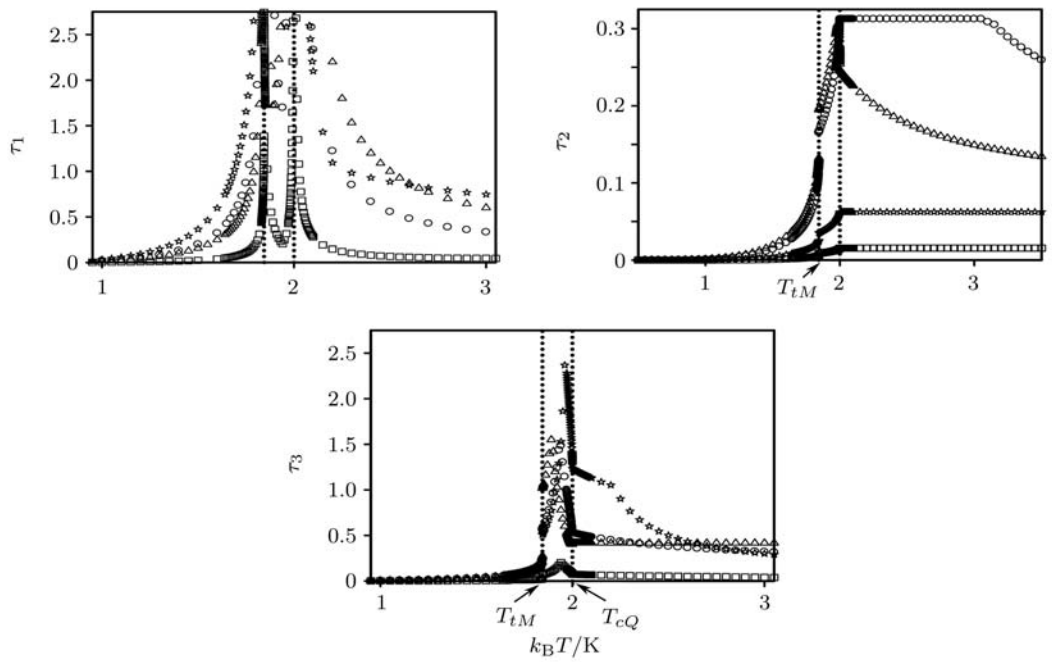


Fig. 2. Behaviours of relaxation times each as a function of reduced temperature for $J/K = 0.53$. The values denoted by symbols are given in Section 4. T_{tM} and T_{cQ} denote the first- and second-order phase transition temperatures for dipolar and quadrupolar moments, respectively.

Thus, we only give here the dependences of the temperature near the first-order phase transitions. The behaviours of τ_1, τ_2 and τ_3 at the first-order phase transition temperatures are presented in Fig. 1. In the figure, the vertical dot lines refer to the phase transition temperature. It is seen from Fig. 1 that all relaxation times remain finite and represent a jump discontinuity at the first-order phase transition temperatures. Therefore, the critical slowdown at the first-order phase transition point is not seen.

On the other hand, for $J/K < 0.46$ the system undergoes two successive second-order phase transitions: the first one is for M (T_{cM}) and the second one for Q (T_{cQ}). The behaviours of τ_1, τ_2 and τ_3 are given in Ref. [27]. However, for $0.46 < J/K < 0.61$ the system also undergoes two successive phase transitions: the first one is a first-order phase transition for M (T_{tM}) and the second one is a second-order for Q (T_{cQ}).[51] We investigated the behaviours of the relaxation times in this case as shown in Fig. 2 for $J/K = 0.53$. τ_1, τ_2 and τ_3 make a jump discontinuity at the first order phase transition point and all of them remain finite, and then if temperature is approaching the second-order phase transition temperature, while one of the relaxation times (τ_1) goes to infinite, the other relaxation times (τ_2 and τ_3) remain finite and make a cusp as seen in the figure. When the system undergoes two successive phase transitions, the behaviours of the relaxation times behave as a single phase transition case at the transition point independent of the next or the previous phase transition type.

It is a conventional way to express the behaviours of these relaxation times near the phase transition points by critical-point exponents. The critical-point exponents are frequently determined by the slopes of log–log plots of data. Although this is a particularly quick method of determining the exponents, it requires a prior knowledge of the critical temperature.[54] Therefore, in order to meet this requirement, we have calculated the equilibrium behaviour previously.[51] We have depicted these relaxation times each as a function of temperature in a log–log plot (see in Fig. 3). Figures 3(a), 3(b), 3(c) and 3(d) show the logarithmic of relaxation times as a function of reduced temperature for a second-order, a first-order, two successive, i.e., first-order and then second-order, and two successive second-order phase transition temperatures, respectively. Figure 3(a) is obtained in the case where the system undergoes a second-order phase transition, small and big marks are for $J/K = 2.5$ and $J/K = 4.0$, respectively. Figure 3(b) is plotted in the case where the system undergoes a first-order phase transition, small and big marks are for $J/K = 0.8$ and $J/K = 1.5$, respectively. Figure 3(c) is depicted in the case where the system undergoes two successive, first- and then second-order phase transitions for $J/K = 0.53$. Figure 3(d) is for the system which undergoes two successive second-orders phase transitions for $J/K = 0.25$.

Since the largest τ_i (τ_1 in this study, which also leads to the slowdown) diverges at the critical point and it is used to determine the dynamic critical exponent z; thus $\tau = \max(\tau_1, \tau_2, \ldots) \sim \xi^z$, where ξ and z are the correlation length and dynamical exponent respectively, hence $\tau \sim |T_c - T|^{-\nu z}$.[55,56] At this point we should mention that for $d > 4$, the critical exponents of the spherical model are the same as those from the mean-field theory, i.e., regardless of the actual value of order parameter (spin) dimensionality, mean-field theory is valid for all models, if $d > 4$. For example, in view of the fact that $\varepsilon \sim \xi^{1/2}$, one can infer that in all regimes of d, the exponent $\nu = \frac{1}{2}\gamma$ and $\gamma = 1.0$.[51,57] Therefore, from Fig. 3, one can see that one of the relaxation times (τ_1) depends on temperature as $(T_c - T)^{-1}$, more precisely, the z dynamic critical exponent is $z = 2 \pm 0.04$, and tends to infinity, while the others depend on temperature as $(T_c - T)^0$ and remain finite near the critical and also tricritical points. However, in the first-order phase transition, several thermodynamical quantities, such as internal energy and order parameter (magnetization), each have a discontinuity, whereas the correlation length is generally finite[52] and the dependences of all relaxation times are given by $(T_c - T)^0$ and remain finite at the first-order phase transition point. Finally, comparing our z value with the z values obtained for different systems experimentally and theoretically, one can see that they are in good agreement with each other. For example, z was found to be 2.1(1) for $FeFl_2$,[58] 2.18(10) for $Fe_{0.9}Zn_{0.1}F_2$,[59] and about 3 for mercury,[60] experimentally, and theoretically; z was also calculated by using damage spreading and heat bath dynamics and found to be 2.172 ± 0.006 for two-dimensional and 2.032 ± 0.004 for three-dimensional spin-1/2 Ising models;[61] by using MC simulations z was found to be 2.62(7) for random Ising model (RIM);[62] by using the Padé–Borel summation technique z was found to be 2.017 for two- and three-dimensional spin systems;[63] by using Chisholm–Borel resummation technique z was found to be 2.012 for

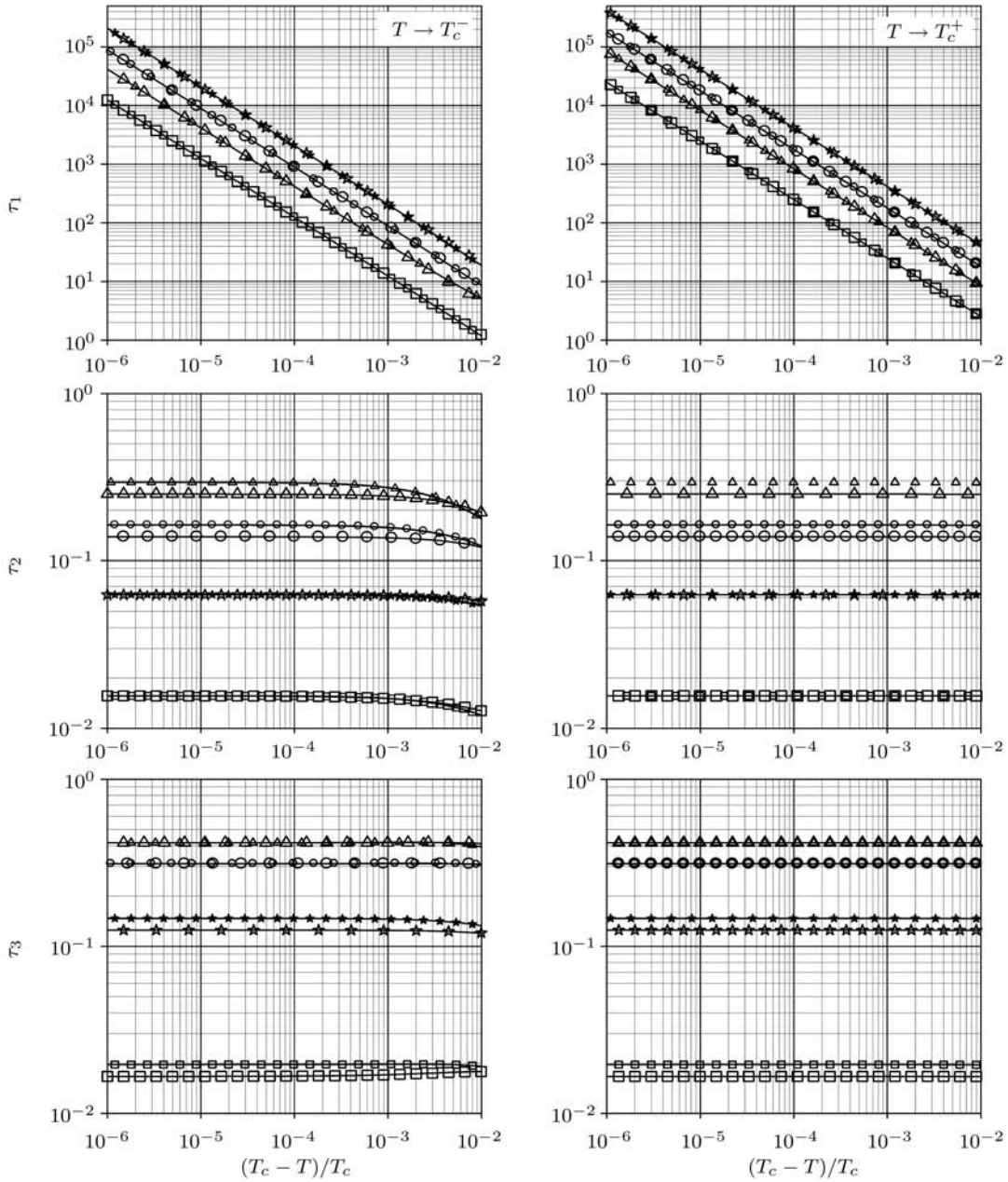


Fig. 3(a). Behaviours of relaxation times each as a function of temperature in a log–log plot. The Onsager coefficients are the same as those in Fig. 1. The small and big symbols are for $J/K = 2.5$ and $J/K = 4.0$, respectively.

three-dimensional magnets with quenched extended defects.[64] For a small site dilution concentration (p), the values of z practically did not differ: $z(p = 0.95) = 2.19(7)$; $z(p = 0.8) = 2.20(8)$.[62,65] Wang and Hu[66] studied the dynamical universality of the spin-1/2 Ising model by using the nonequilibrium relaxation (NER) method and estimated the value of z to be 2.166(7), 2.164(7) and 2.170(10) for square, triangular and honeycomb lattices, respectively. Wang[67] studied the NER process of the two-dimensional spin-1/2 Ising model with the series expansion method, and obtained $z \approx 2.170$. Wang also showed that the combination of the series expansion and the MC simulation provided $z = 2.169(3)$. Murase[68] studied the NER in three-state Potts models on square, honeycomb and triangular lattices weparately and found z to be 2.20(1).[69] Miyashita and Takano[70] investigated the scaling property of the relaxation time in the vicinity of the critical temperature and found the critical exponent z to be about 2.2 for the two-dimensional Ising model. From time relaxation of the magnetization and energy of the Ising model, Ito[71] found that $z = 2.165 \pm 0.010$ for the square lattice Ising model, which was also confirmed by other calculations.[61,72,73] As a summary, estimates of z varied in a

large range between 1.7 and 2.3. On the other hand, z was found to be 3 instead of 2 for conserving dynamics, recently.[74]

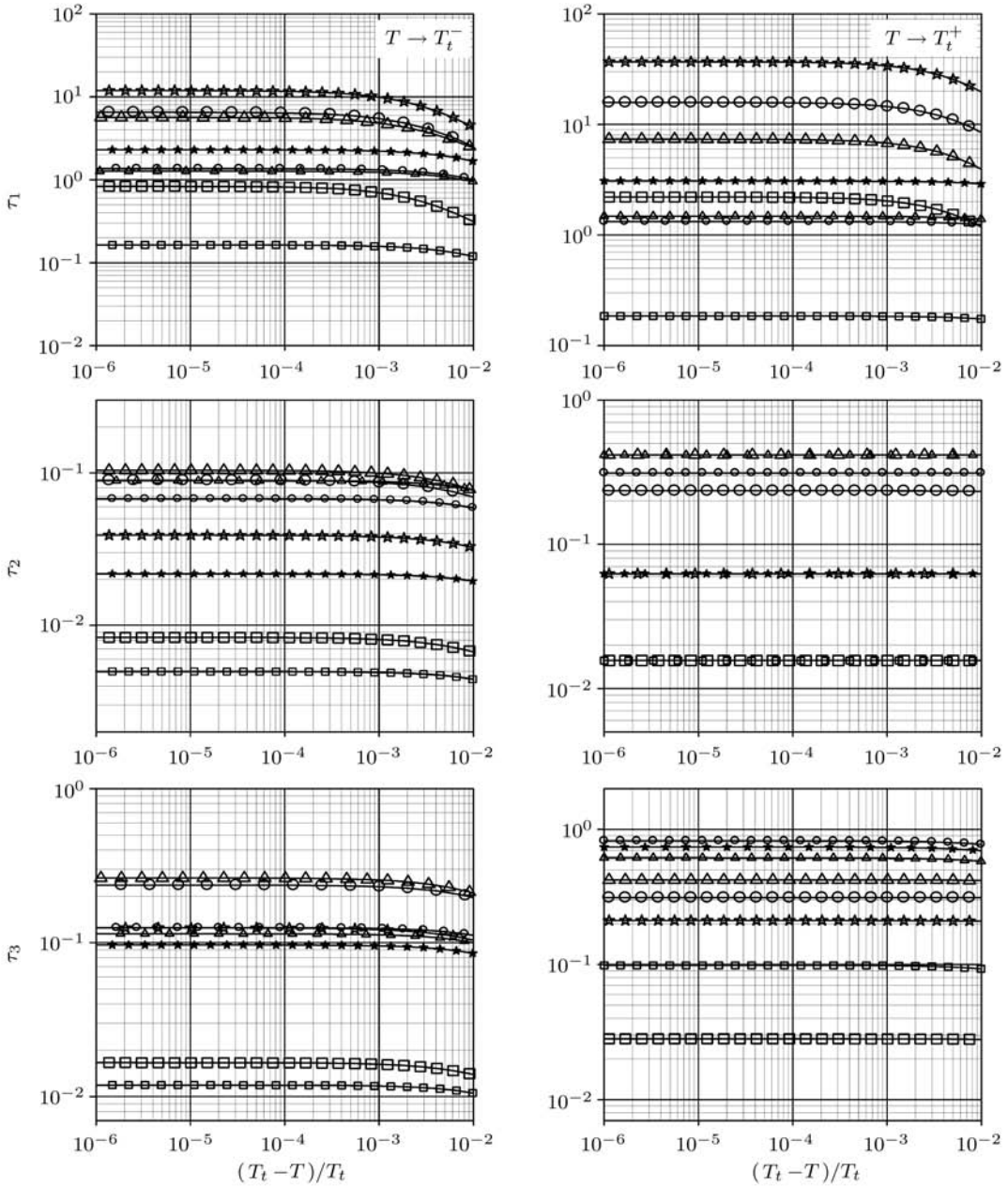
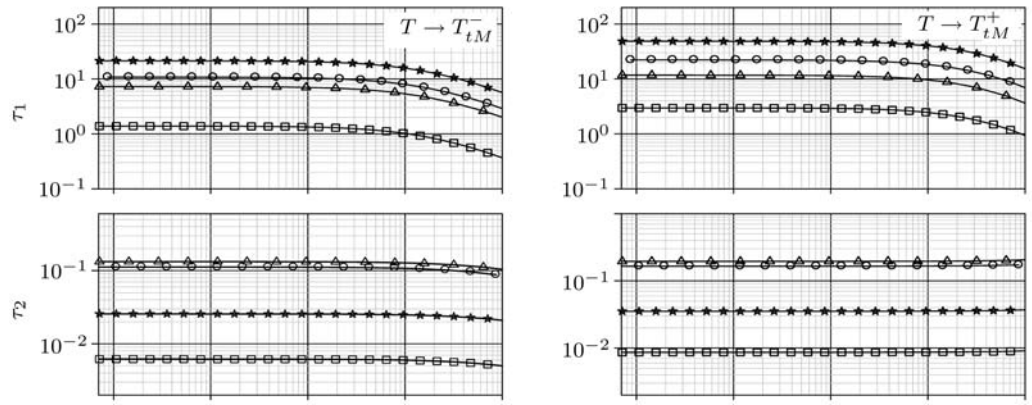


Fig. 3(b). The same as in Fig. 3(a) but for $J/K = 0.8$ and $J/K = 1.5$, respectively.



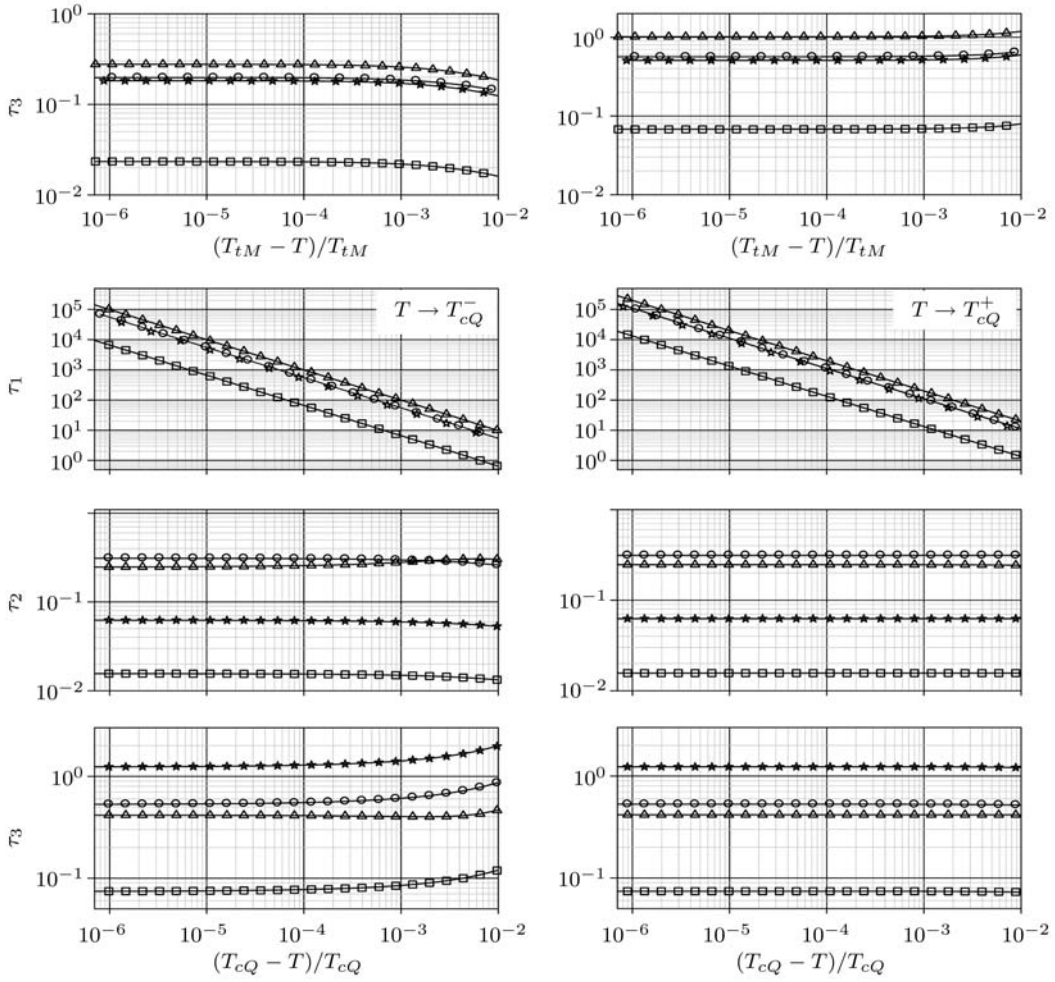
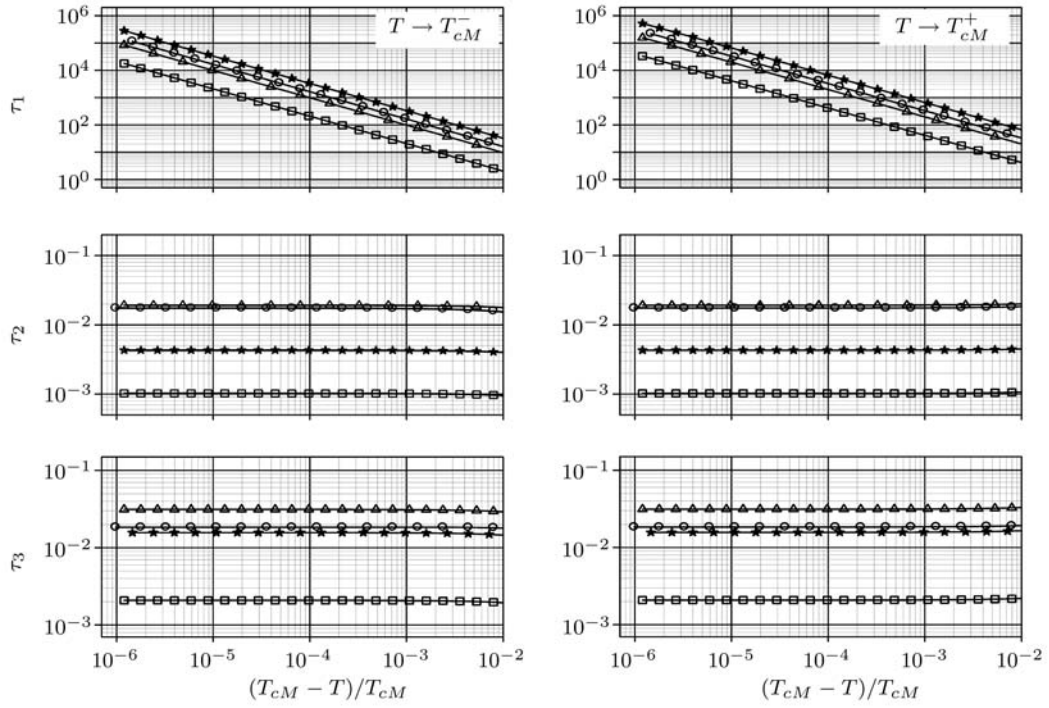


Fig. 3(c). The same as in Fig. 3(a) but for $J/K = 0.53$.



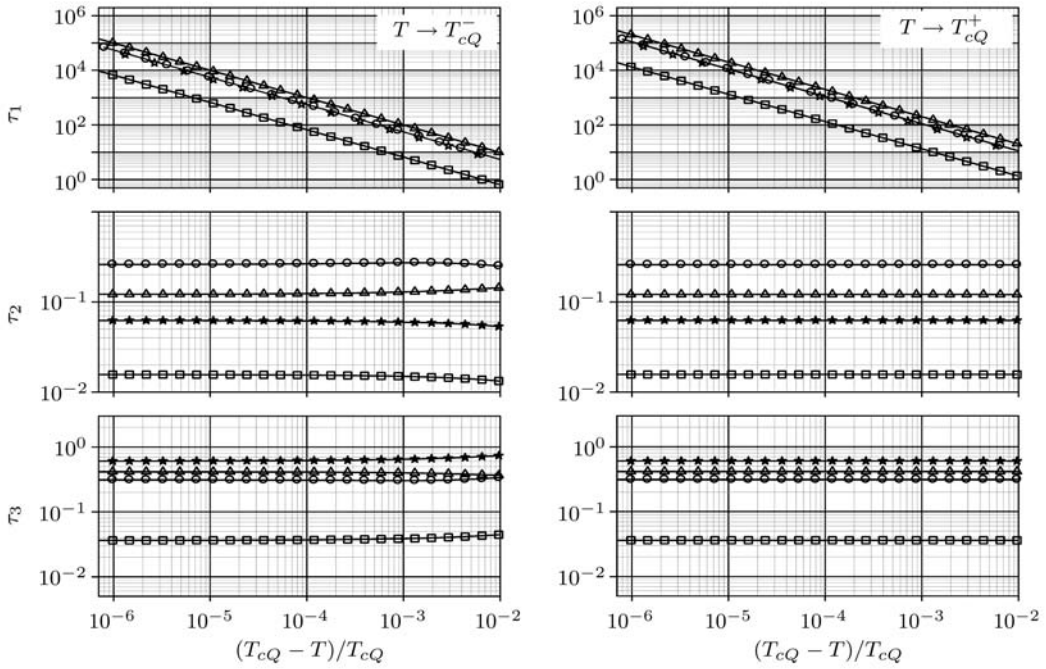


Fig. 3(d). The same as in Fig. 3(a) but for $J/K = 0.25$.

5. Summary and conclusion

In this paper, we studied the relaxation behaviours of the order parameters of the isotropic spin-3/2 BEG model by means of Onsager's theory of irreversible thermodynamics. More specifically, using an expression for the free energy obtained in the molecular-field approximation in the Bragg–Williams formalism, the entropy produced in the irreversible process is calculated and the time derivatives of order parameters are treated as fluxes conjugate to their appropriate forces in the sense of Onsager's theory. The kinetic equations are obtained by introducing the phenomenological kinetic coefficients that satisfy Onsager reciprocal relations. The solution of these equations near equilibrium states is given by three relaxation times which describe the nonequilibrium behaviour in the cooperative system. The behaviours of these relaxation times each as a function of temperature are determined and examined for temperatures near the phase transition points.

We find that as expected,[52] one of the relaxation times (τ_1) increases rapidly with temperature increasing and tends to infinity near the critical and tricritical points as $(T_c - T)^{-1}$, more precisely the critical exponent is $z = +2 \pm 0.04$, which is in a good agreement with the z values obtained in different systems, experimentally[58−60] and theoretically,[61−73] but z is found to be 3 instead of 2 for conserving dynamics, recently.[74] On the other hand, τ_1 makes a jump discontinuity at the first-order phase transition point. The other relaxation times scarcely vary with temperature and they increase slightly just below and above the first- and second-order phase transition temperatures and also the tricritical point; hence the critical exponent is zero, namely $(T_c - T)^0$. Moreover, the other two relaxation times make a cusp at the critical and the tricritical points, but they make a jump discontinuity at the first-order phase transition temperature.

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